Pergamon

Chemosphere, Vol. 37, Nos 9-12, pp. 2173-2197, 1998

0045-6535/98 $19.00+0.00

PII: S0045-6535(98)00279-3

RELATIONSHIPS BETWEEN DIOXINS IN SOIL, AIR, ASH, AND EMISSIONS FROM A MUNICIPAL SOLID WASTE INCINERATOR EMITTING LARGE AMOUNTS OF DIOXINS

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ABSTRACT

The Columbus Municipal Waste-to-Energy (Columbus WTE) facility in Columbus, Ohio, began operation in June, 1983 and ceased operation in December, 1994. During its operation, it was estimated to have released nearly 1,000 grams of dioxin Toxic Equivalents (TEQs) per year. This compares to a 1994 estimate of 9,300 g TEQ/yr from all sources emitting dioxins into the air in the United States (EPA, 1994), and to total releases of dioxins near or below 1,000 grams TEQ/yr for England (Eduljee and Keyke, 1996), Belgium (Wevers and De Fre, 1995), and West Germany (Fiedler and Hutzinger, 1992). Because of the magnitude of emissions from this single source, studies were undertaken to evaluate the impacts to air and soil near the incinerator. This paper presents analyses evaluating dioxin concentrations and profiles in four media: stack gas, ambient air within 3 km of the incinerator, soil samples up to 8 km from the incinerator, and incinerator ash. Principal findings include: 1) an "incinerator signature" profile, as defined by stack gas emissions, was found in the ash and in subsets of the air and soil matrices, 2) soil concentrations declined from directly outside the incinerator property to the city at large, 3) an urban background soil concentration of dioxin Toxic Equivalents (TEQs) was estimated at 4 pg/g, while concentrations generally within 2 km of the incinerator ranged from 4-60 pg TEQ/g, 4) an urban background air concentration was estimated at 0.05 pg TEQ/m^3, while air concentrations at a specific location about 2 km in the downwind direction of the incinerator had concentrations of 0.17 and 0.35 pg TEQ/m^3 during two sampling dates, 5) analysis of the soil monitoring data in combination with the stack test data suggests that less than 2% of emitted dioxins can be found in the soil near the incinerator, and

6) principal component analysis suggests that the fraction of total concentration of OCDD is the single feature explaining most of the variation of all concentration profiles. This paper discusses these and other findings, and their implications.

Introduction

A stack test conducted in 1992 indicated unusually high emissions of dioxins from the Columbus Waste to Energy (WTE) municipal solid waste incinerator. The Solid Waste Authority of Central Ohio (SWCO) attempted to reduce emissions with a series of process modifications. A second stack test taken in March, 1994, showed a four-fold reduction in emissions as compared to the 1992 stack test. Meanwhile, the Ohio Environmental Protection Agency (OEPA) used air monitoring to evaluate air quality in the city of Columbus. A first round of monitoring occurred in March and April of 1994 (OEPA, 1994b). In November of 1994, SWACO announced the planned closure of the Columbus WTE on December 31 of 1994. In the summer of 1995, the OEPA performed a second round of ambient air monitoring to evaluate the air quality now that the incinerator was no longer operating (OEPA, 1995).

In 1995, a workgroup comprised of individuals from the United States Environmental Protection Agency (USEPA), the OEPA, the Ohio Department of Health, and the Agency for Toxic Substances and Disease Registry discussed approaches to evaluating the environmental impact of emissions from the Columbus WTE. Ambient air monitoring was no longer an option, since the Columbus WTE was shut down, and vegetation and animal products (milk, e.g.) were also rejected based on evidence that the principle pathway for dioxins to enter the food chain was from the air to the plant to the animal (EPA, 1994). The group decided that soil was the best matrix to evaluate long term impacts. A first round of soil samples was taken in December of 1995 and a second round was taken in August of 1996.

The final set of measurements used in the analyses of this paper were measurements of incinerator ash concentrations taken in 1987 as part of a Superfund site investigation (EEI, 1987).

The purposes of the analyses in this paper are as follows: 1) to ascertain relationships between the four matrices (stack gas, ash, soil, ambient air), if they exist, 2) to determine any long and short term impacts from operation of the facility, and 3) to determine background soil and background ambient air concentrations for the city of Columbus, if possible. A preliminary analysis of some of the data in this paper can be found in Lorber, et al. (1996a,b).

Description of Data

During its 11.5 years in operation (6/83-12/94), the Columbus WTE facility processed an average of 1,600 metric tons of solid waste per day. The facility had three stacks and six boilers. A

stack test, conducted in 1992, entailed five samples from one of the three operating stacks. The average total (sum of all Cl4-Cl8 homologue groups) and Toxic Equivalent (TEQ) concentrations in the stack gas were 6,799 ng/m^3 and 136 ng TEQ/m^3, respectively. Extrapolating these test results to "normal operating conditions", (i.e., number of boilers continually operating, volumes of air emitted, etc.), the OEPA estimated that emissions of dioxin TEQs would equal $3.12*10^{-5}$ g/sec (OEPA, 1994a), which translates to an annual emission of 985 g TEQ. Process modifications were undertaken in the winter of 1993/94 in an attempt to reduce dioxin emissions. A stack test conducted in March of 1994, which entailed three samples from one of the stacks, showed reductions in concentrations to 3,685 ng total/m^3 and 64 ng TEQ/m^3. Reductions also occurred in stack emission air volumes, leading to an estimate of annual emissions of 267 g TEQ. Further details on the 1992 stack test can be found in EERG (1992), and details from the 1994 stack test can be found in EMC (1994). A total of 8 stack gas samples, 5 in 1992 and 3 in 1994, were available for this analysis.

Ambient air monitoring was conducted by the OEPA in 1994 while the incinerator was operating (but after process modifications reduced emissions of dioxins), and in 1995 after the incinerator shut down. General Metal Works model PS-1 high volume samplers were used to collect 48-hr samples. Concentrations were, therefore, the sum of vapor + particle phase concentrations. Six monitors were in the city of Columbus between 1.8 and 3.0 km from the site, mostly in the historical downwind direction, northeast, but one in the upwind southwest direction. A seventh sampler was located 45 km southwest of the facility in a rural "background" setting. Six samples were taken each in March and April, 1994, and seven samples were taken in June of 1995. The March set, taken on the 15-17th of the month, occurred at nearly the precise time that the March 1994 stack testing occurred, on the 16-18th. In all, there were 10 urban air samples taken during 1994 (5 on each of two sample dates), 6 urban samples in 1995, and 3 background samples (2 in 1994 and 1 in 1995), for a total of 19 air samples. Wind rose data were also available for the 48-hr periods of both air sampling dates in 1994, thereby allowing for additional observations on the likelihood of certain samplers being impacted, or not impacted, by emissions from the Columbus WTE. Full details on the air monitoring studies, including analytical methodologies, quality control, and final results, are described in OEPA (1994b, 1995).

A first phase of soil sampling was conducted by USEPA in December of 1995 (EPA, 1996). Sampling in this round included 4 samples on the site of the incinerator, 18 samples within about 3 km of the incinerator in the city of Columbus, and 3 samples at a background site 45 km away. This background site was the same as the air monitoring background site. The study design for this phase employed a stratified random selection process, involving sites in the four major quadrants around the incinerator (northeast, southwest, etc.) with an emphasis of sampling in the quadrant

which was historically downwind from the incinerator, the northeast quadrant. The following conditions were sought during site selection: 1) level, undisturbed soils, 2) away from trees, 3) not adjacent to roads, 4) not near pressure treated wood, and 5) not known or suspected to have high dioxin concentrations for any other reason. All samples were collected using pre-cleaned equipment dedicated to each sampling location. Each sample site consisted of an area of 1.5 m x 1.5 m. A grid of 25 sections was established at each site and used for random selection of aliquot sample sites. Four random aliquots were collected for each sample. A "sample" for this study was, therefore, a composite of four aliquots. Aliquots were collected using a stainless steel tulip bulb planting device. This device removed a plug approximately 7.5 cm in diameter to a depth of about 7.5 cm.

A second phase of soil sampling was undertaken in August of 1996. Thirteen samples were taken from about 2 km away from the incinerator to about 8 km distant. The purpose of this second round was to ascertain whether a background concentration for the city of Columbus could be determined. A similar selection criteria for sample sites was employed in this second round.

Altogether, there were 4 soil samples on the incinerator property, 31 samples in the city of Columbus taken from right outside the incinerator to upwards of 8 km away, and 3 background samples taken 45 km away, for a total of 38 soil samples.

Figure 1 shows the location of the incinerator in relation to the 32 soil samples in Columbus and the 6 urban air samplers. Not shown in this figure are 3 of the 4 soil samples taken on the site of the incinerator, and the background site in which 3 soil and 3 air samples were taken. This figure identifies the groupings of the soil samples, as described in the results section below.

Five samples of ash were measured for dioxins and furans as part of a Superfund site investigation in 1987 (EEI, 1987). Two ash samples were combined bottom and top ash, one was a bottom sample, one a top sample, and the last was a stack scrape sample. These ash samples were the final matrix for analysis.

Methods of Interpretative Analysis

The magnitude of the dioxin concentrations was the most straightforward measure in evaluating and comparing the different matrices. "Total" concentrations in this paper are defined as the sum of the dioxin and furan homologue Cl4 -Cl8 group concentrations. Toxic Equivalent, or TEQ, concentrations are also discussed. A TEQ concentration comprised of the 17 dioxin and furan congeners (abbreviated CDD/F) is determined using the International Scheme (EPA, 1989).

A second way to evaluate and compare the different media is through the use of "congener profiles". These describe the relative concentrations of the congeners among each other. There is not a uniform approach to defining CDD/F congener profiles. Hagenmaier, et al. (1994), for

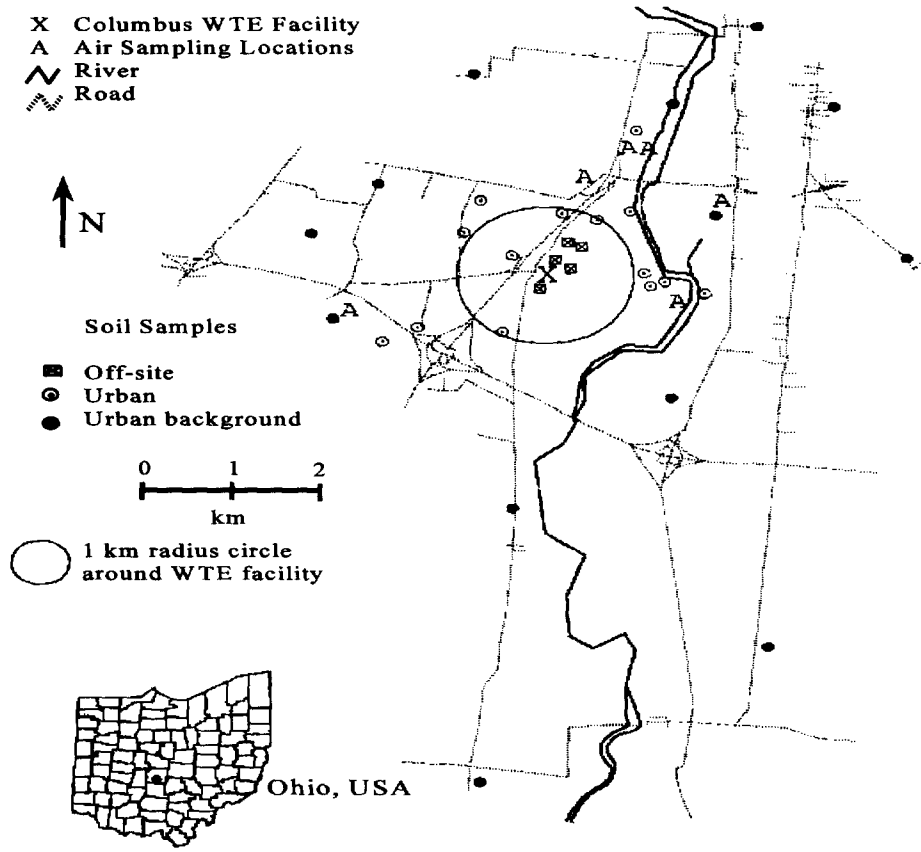

Figure 1. An overview of the study area showing the location of the Columbus WTE, the soil samples, and the urban air samplers.

example, defined a "characteristic profile" of a given source of dioxin release or environmental media, by dividing each congener's concentration by the concentration of its respective homologue group. For example, a characteristic fraction for 2,3,7,8-TCDD would be calculated as the concentration of 2,3,7,8-TCDD divided by the concentration of the homologue group, TCDD. In the preliminary analysis of the data in this paper, Lorber, et al. (1996a,b) added the concentrations of all toxic congeners to get the "total" concentration, and then divided each congener's concentration by that total. Of course, that "total" concentration is different than the "total" concentration defined here (the sum of the Cl4-Cl8 homologue group concentrations).

In this paper, two types of profiles are defined. Homologue profiles are defined as a vector, $h_1,h_2,...h_{10}$, where each h_i is the concentration of the given homologue group divided by the total

concentration. Congener profiles c_1, c_2, .. .c_{17}, are similarly constructed with each congener concentration being divided by the total concentration. Since these profiles are vectors of high, 10 or 17, dimension, and therefore problematic to analyze, standard methods that effectively reduce the dimensionality of this data are used. One approach is principal components analysis, which attempts to reduce the dimension of a vector by identifying linear combinations which explain a high proportion of the total vector variance. The other approach is to create a distance metric that calculates the "distance" between two profiles. Distance can be defined as the usual Euclidean distance in the appropriate dimensional space or in some other fashion.

For one analysis in this paper, a non-linear regression model was fit using maximum likelihood where the error was assumed to be normally distributed with standard deviation proportional to the mean value. Significance testing was done using the likelihood ratio test.

Arc/Info® (Version 7.0.3), which is a Geographical Information Systems (GIS) software package, was used to derive soil concentration contours. The kriging option in Arc/Info, using the default spherical semi-variogram model, created a lattice with a spacing of 85 meters. Then, the lattice contour option of Arc/Info was used to draw the contour lines.

Finally, it is noted that all average concentrations reported in this paper were calculated assuming non-detects were equal to ½ detection limit (unless otherwise noted). A separate analysis showed that there was only a marginal difference in concentrations if non-detects were instead assumed to be 0.00.

Results

The results from this paper will be described in a series of principal findings, which are italicized and indented, followed by the supporting evidence for that finding.

The fractional contribution of OCDD to the homologue or congener profile was the important distinguishing feature in different groupings of media samples.

There were a total of 70 samples analyzed for dioxin concentrations, including 38 soil samples, 19 air samples, 8 stack samples and 5 ash samples. Principal component analysis was performed on both the homologue and the congener profiles using data from all 70 samples. This analysis showed that a high percentage of the total variance of the profiles could be explained by the first principal component, and that this first component was made up largely of the variable representing percentage OCDD. Taken together, these results showed that 79% of the total homologue profile variance and 84% of the total congener profile variance could be explained by the variance of the OCDD percentage alone.

Analysis of the profiles using the Euclidean distance metric supports the above findings of

the importance of OCDD. Using this metric, the distances between all air, soil and ash samples and each of the 8 stack samples were computed; these distances were alternatively computed using just the absolute difference in OCDD percentage. These two distance measures, the multidimensional one based on the entire profile, and the simple one based simply on OCDD, were found to be very highly correlated, with r = 0.99 for homologue profiles and r = 0.92 for congener profiles.

Taken together, these results suggest that for these data, the percentage OCDD is an acceptable surrogate for the entire homologue or congener profile. Thus, in the subsequent findings, the OCDD percentage is the principal metric used to compare profiles. In some cases, the entire profile will be examined for the sake of completeness.

An "incinerator signature" was identified by the stack emission profile, and was found in the other three media: ash, soil, and air.

An "incinerator signature" is traditionally identified by the profile of the stack emissions. The Columbus WTE signature was defined as the average of the congener profiles determined from the 1992 and 1994 stack gas tests. Specifically, a unique profile was determined for each of the two stack tests as the average concentrations in the samples taken for each test (5 in 1992 and 3 in 1994), and then the signature was calculated as the average of the two profiles, equally weighted. It was found that very similar signatures were in the ash samples, and in subsets of the soil and air samples.

Figure 2 shows the stack gas signature profile and the average profiles of subsets of the other three matrices that had the same signature. OCDD is the most prominent congener for the air, ash, and soil matrices, and nearly the most prominent congener in the stack gas. All profiles are also characterized by relatively high levels of OCDF, 1234678-HpCDF and 1234678-HpCDD. Further, for all four of these groups, there were a number (ranging from 4-8) of other congeners with percentages above 1%. The profiles for these groups are easily distinguished from those of the urban background and rural soil and air samples, where OCDD is much higher and only OCDF, OCDD, 1234678-HpCDD and 1234678-HpCDF make up more than 1% of total homologue concentration. Observations on background congener profiles in soil and air are presented in a subsequent finding.

The ash, soil, and air "incinerator signature" subsets, are further described:

a) Ash: The average total concentration of the five ash samples is 608 ppb (ng/g or 1,000 times higher than the ppt, or pg/g, units used otherwise throughout this paper for soil concentrations), and the average TEQ concentration is 14.6 ppb. The stack scrape sample had the highest total and TEQ concentrations at 1,296 and 28.8 ppb, respectively. The ash sample having the lowest concentration was the sample identified as bottom ash, which had mostly non-detects.

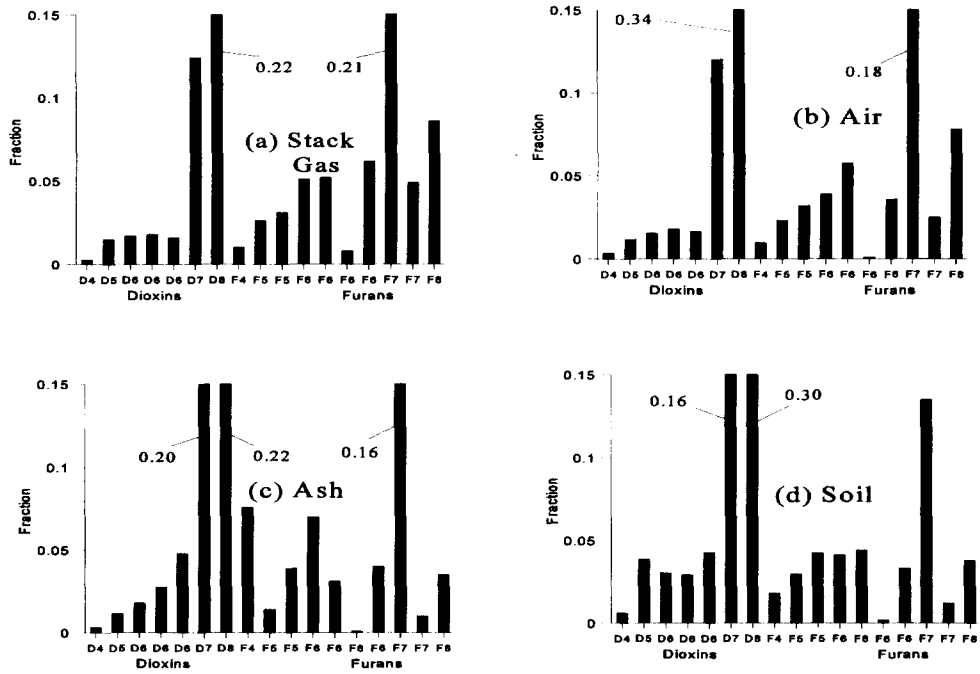

Figure 2. Congener profiles for the Columbus WTE incinerator signature including: (a) stack emissions averaging 1992 and 1994 profiles, (b) two downwind air concentrations, (c) average of 5 ash measurements, and (d) average of 3 on-site soil concentrations (note: D4 = 2,3,7,8-TCDD; D5 = 1,2,3,7,8-PCDD; order of congeners on x-axis matches that of Tables 1 and 2).

The detection limits were not listed, and the congeners and homologue groups which were detected had concentrations above 30 ppt. Therefore, the calculation of averages here with ND = 0 did not change the basic message.

b) Soil: Of four soil samples taken on-site, it appeared that three of them would be part of this group based on their concentrations alone. The average total and TEQ concentrations of these three samples were 15,747 ppt and 458 ppt, respectively. In contrast, the fourth on-site sample had total and TEQ concentrations of 1,199 ppt and 50 ppt, respectively. Two of the high soil concentrations were taken near the ash storage pits, and the third was adjacent to one of two roads leading to and from the incinerator. Among other uses, this road was used to transport the ash to landfills. The second road led to the employee parking lot. The fourth, much less impacted, soil sample was taken on open grounds adjacent to this parking lot. It would appear that the two soil

samples of this study taken near the ash pits, along with the soil sample near the ash transportation route are similarly impacted by ash spillage or drift, rather than incinerator emission depositions. If depositions were the cause of such high concentrations, than it seems logical that the fourth on-site sample would be similarly impacted. Also, none of the soil samples, some immediately off-site, are nearly this high in concentration.

c) Air: Recall that the Columbus WTE was operating in 1994 when the air monitoring occurred. The maximum air concentrations found in the 1994 air monitoring were from the same monitor located about 2 km from the WTE site in the easterly direction. The concentrations found there on the two 1994 sampling dates were 9.2 and 19.0 pg/m^3 total, and 0.17 and 0.35 pg TEQ/m^3, respectively. Although this direction was not quite the historical downwind direction of southwest to northeast, the wind rose data presented in OEPA (1994b) showed that this was the predominant downwind direction during these sample dates. OEPA (1994b) also noted that these two high air concentrations were located downwind of the Columbus WTE.

> *There is a clear gradient in the soil results, with declining concentrations as a function of distance from the incinerator. An examination of the data suggests that the urban background soil concentration is about 530 ppt total and 4 ppt TEQ.*

The 38 soil samples of this study are grouped into five clusters as follows: 1) 3 "on-site" samples taken on the site of the Columbus WTE. As discussed above, these were likely the result of ash transport, spillage, or drift, 2) 5 "off-site"samples, which include one of the samples taken on the site of the Columbus WTE, near the employee parking lot as described above, and 4 in the historically northeast direction within 1 km of the facility, 3) 14 "urban" samples in the city of Columbus which were within about 3 km of the plant and were part of the first phase of soil samples, 4) 13 "urban background" samples which were the second phase of soil samples taken from about 3 to as far away as 8 km from the Columbus WTE but still within the city of Columbus, and 5) 3 "rural background" samples taken at a site 45 km away. Table 1 shows the average concentrations of the 17 toxic congeners, the 10 homologue groups, the total and the TEQ, for the five groups of soil samples.

Before commenting on results from this table, it is noted here that a total of 5 samples taken in this soil monitoring program were not included in this table. There were 2 soil samples taken in the December 1995 sampling that were unusually high and the laboratory recoveries were below quality assurance standards (i.e., less than 40% for the majority of the congeners). The total concentrations (i.e., sum of the homologue group concentrations) of these two samples were about 13,000 ppt and 24,000 ppt. These compare to all other urban soil samples which were at about 5,000 ppt and below. Only the soil samples on the Columbus WTE, averaging about 16,000 ppt,

Table 1. Congener-specific, homologue, and TEQ concentrations for the five clusters of soil samples.

Congener	On-site (n=3)	Off-site(n=5)	Urban (n=14)	Urban Background (n=12)	Background (n=3)
2378-TCDD	37	4	2	0.6	0.4
12378-PCDD	233	18	3	1.3	0.1
123478-HxCDD	183	17	3	1.2	0.4
123678-HxCDD	178	24	6	2.0	0.8
123789-HxCDD	259	28	6	3.1	1.2
1234678-HpCDD	973	395	112	32.5	18
OCDD	1776	3245	892	344.7	161
2378-TCDF	111	10	2	1.1	0.5
12378-PCDF	179	15	2	1.1	0.2
23478-PCDF	257	22	4	1.7	0.2
123478-HxCDF	250	32	4	2.0	0.2
123678-HxCDF	268	25	4	1.9	0.5
123789-HxCDF	15	1	0.3	0.2	0.2
234678-HxCDF	200	20	3	2.0	0.6
1234678-HpCDF	819	106	23	10.8	4.0
1234789-HpCDF	74	10	1	0.6	0.3
OCDF	231	63	26	8.8	11
TCDD	1122	102	15	4	1
PCDD	1832	73	12	12	0.3
HxCDD	1907	172	51	22	5
HpCDD	1726	714	203	60	38
TCDF	2158	164	37	11	2
PCDF	2579	218	40	24	2
HxCDF	1214	135	30	19	3
HpCDF	1202	212	60	19	12
PCDD/PCDF	**15,747**	**5,132**	**1,360**	**528**	**235**
TEQ	**458**	**49**	**10**	**4**	**1.4**

were comparable to these two sites. Both sites were revisited for the second round of sampling in August of 1996. One of the sites continued to show the same very high soil concentrations, but the second sample showed low concentrations, typical of background conditions, indicating possibly

that the exact same spot from the first round was not found. The judgement was made that the original two 1995 soil samples may have been influenced by activities not related to emissions from the Columbus WTE. For this reason, and also because the two samples failed laboratory quality control measures, they were rejected from the analysis of this paper. The resamples of these two sites were also not included in the analysis, since they were not part of the study design, but rather were taken only to evaluate the locations sampled in the first phase. The fifth rejected sample came from the urban background group of 13 samples. One sample was clearly an outlier in that it had a total concentration of 7,900 ppt. Among the other 12 "urban background" samples, the high total concentration was 1,590 ppt. The high total concentration in the outlier sample was primarily OCDD; the OCDD level was 7,570 ppt. No explanation could be found for this high OCDD reading at this anomalous site, although it should be noted that several other urban background soil samples had OCDD concentrations above 1,000 pg/g. All further analyses were performed excluding this sample. The urban background set had a final total of 12 samples, and the final total of all soil samples was reduced from 38 to 37.

As seen in Table 1, the average total and TEQ concentrations in a group decreases as the average distance of the group from the WTE site increases. The urban background soil concentration is about 530 ppt total and 4 ppt TEQ, while the rural background soil concentration is about 230 ppt total and 1.4 ppt TEQ. Two other clusters of soil samples off the site but within 2 km of the incinerator had concentrations elevated in comparison to this urban background - the concentrations in the nearest cluster averaged about 5,100 pg/g total and 49 pg/g TEQ, and in the further cluster averaged about 1,400 pg/g total and 10 pg/g TEQ.

The decline in concentrations as a function of distance is steeper for TEQ concentration as compared to total congener concentration. This pattern can be explained by the fact that, as seen in Table 1, the average OCDD concentrations decrease less rapidly with distance than do the concentrations of the other congeners and homologues. OCDD, because of its low toxic equivalency factor of 0.001, influences the TEQ only minimally. Thus the effect of a smaller decrease in OCDD would be seen most in the total congener concentration and least in the TEQ concentration.

To further explore the relation between total concentration and distance from the site among the urban samples, non-linear regression utilizing the following simple model was performed:

$$y_i = C_o e^{-r\,(distance)} + BACKGROUND + e \qquad (1)$$

which postulates that the difference between the total concentration in soil, y_i, and the "urban" background total concentration decreases exponentially with the distance of the soil from the WTE

site. Here the "urban" background concentration is estimated by the parameter BACKGROUND, the parameter r reflects the rate at which concentration falls off with distance, the parameter C_0 gives the concentration above background at 0 distance from the site, and e is the mean error.

This model was run both with and without the three "on-site" samples since, as speculated above, ash management rather than stack emission deposition may be responsible for these samples. The results of the model showed that adding the exponential decay term significantly improved the fit of the model (both with and without the on-site samples) over a model with just a background (i.e., intercept) term. The inclusion of the 3 onsite samples did not appreciably alter the model equations. However, based on the speculation that these soil results were influenced by ash management, only the results excluding these samples were pursued further. The final equation was 13,000*exp (-1.75[distance]) + 514, where distance was in kilometers, and the 13,000 and 514 are in ppt units. It is perhaps not coincidence that this modeled *BACKGROUND* concentration of 514 ppt is very close to the average total concentration in the "urban background" group of 528 ppt (see Table 1). The equation indicates that soil at distances of, say, 0.6, 1.8, and 3.2 km from the WTE site would have (on average) concentrations about 10 times, 2 times, and 1.1 times the background concentration, respectively. The results of this analysis are shown in Figure 3, which displays the concentration of the 31 soil samples (37 total minus 3 on-site samples minus 3 rural background samples) as a function of distance from the incinerator, including the curve-fit equation.

The predominant wind direction is from the Southwest to the Northeast. Therefore, one would expect the highest soil concentrations to be Northeast of the incinerator. This trend was verified with kriging.

The trend of this finding is displayed graphically in Figure 4, which was generated using the kriging option of Arc/INFO (as described in the methods section above). This figure was generated with only 19 of the 37 soil samples of this study: the 5 "off-site" samples and the 14 urban samples. The spatial density of the urban background and rural samples was too low for reliable kriging, while the 3 on-site samples were thought to be affected by ash. As seen in this figure, lines of equal TEQ concentration extended in concentric rings around the incinerator. The isolines of 20, 30, 40, and 50 ppt TEQ extend furthest in the Northeast direction, which, as noted above, is also the predominant wind direction (OEPA, 1994).

The "off-site" urban soil cluster had total (5,132 ppt) and TEQ (49 ppt) concentrations that were distinctly elevated from other soil clusters. The most likely source of dioxins in the soil characterized by this cluster are emissions from the Columbus WTE (with perhaps drift

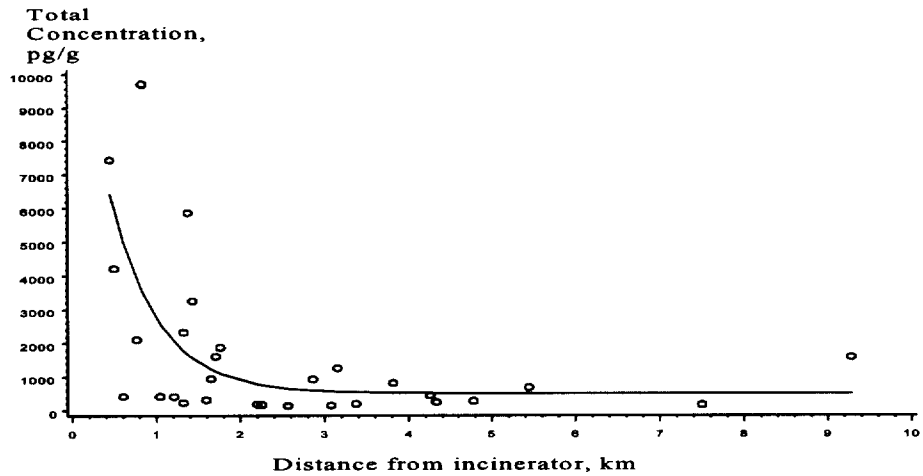

Figure 3. The relationship of soil concentrations of dioxins with distance from the Columbus WTE.

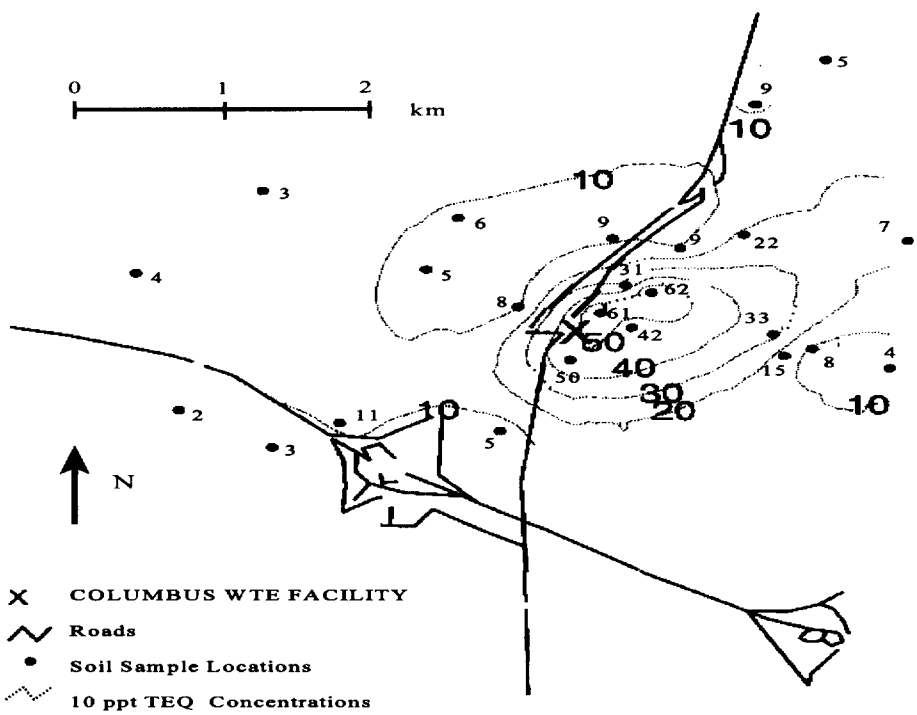

Figure 4. An overview of the area of soil impacted by depositions from the Columbus area, including the soil TEQ concentrations at sample points and a delineation of 10 ppt TEQ contours.

of ash as a possible source; no other sources were identified). However, the profile in this cluster is distinctly different than the "incinerator signature" profile described earlier, and in fact, remarkably similar to all other soil cluster profiles. If the dioxins in this off-site cluster can be attributed only to incinerator emissions, than this is evidence that dioxins in emissions undergo transformations, either in the air before reaching the soil, or within the soil itself, to result in a soil profile that is distinct from the profile in the stack emissions.

Figure 5 shows the congener profile of all four soil clusters; the fifth cluster, the "on-site" cluster, is not included. As easily seen, they are remarkably similar, with OCDD fractions around 0.65. I n contrast, the average OCDD fraction in the on-site group is 0.11 (range of 0.10 to 0.12). This fraction is still clearly higher than the average OCDD fraction in the 1992 stack samples of 0.03. It is, however, more in line with the 1994 stack emission test, where the OCDD fraction averaged 0.17 (range of 0.13 to 0.21). Among the four clusters of soil samples outside the incinerator, there may be a slight trend in an increasing OCDD fraction as a function of distance.

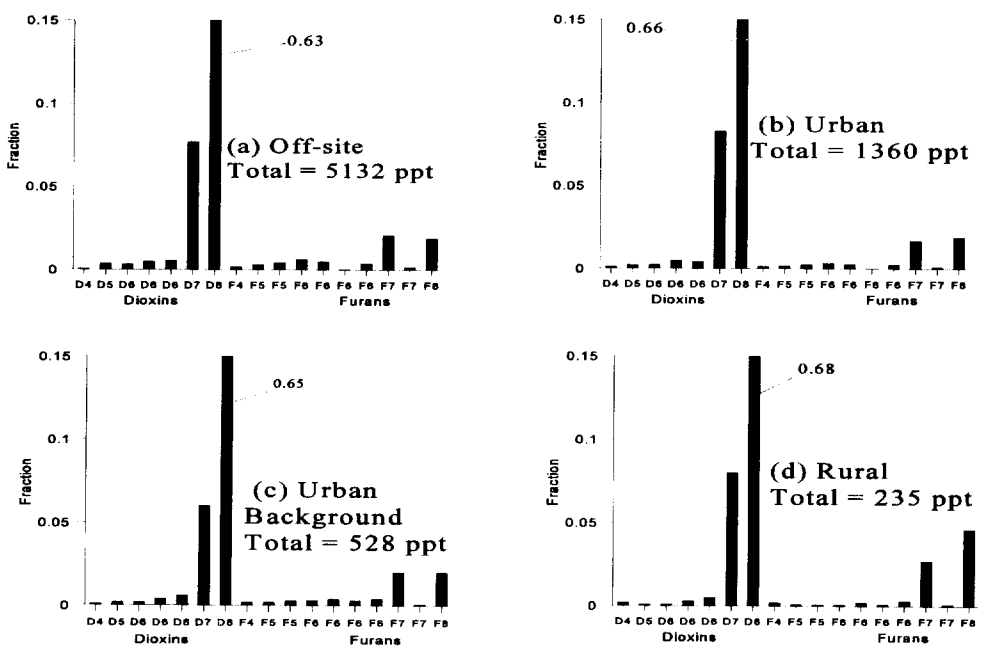

Figure 5. Congener profiles for the four clusters of soil samples outside the incinerator property including, (a) the off-site cluster, (b) the urban cluster, (c) the urban background cluster, and (d) the rural cluster (note: D4 = 2,3,7,8-TCDD; D5 = 1,2,3,7,8-PCDD; order of congeners on x-axis matches that of Tables 1 and 2).

For the "off-site" group, the OCDD fraction is 0.63. The urban and urban background groups have essentially equal fractions of 0.65. In the rural setting 45 km away, the OCDD fraction is 0.68.

With no other sources in the vicinity identified, it would appear that stack emissions explained the high soil concentrations in the off-site cluster. It may be possible that ash drift also contributed to these soil concentrations, but there is no evidence of this, and no reason to believe that it would occur. Most noteworthy, as indicated, was that the profile in this off-site cluster does not resemble the stack emission profile. This finding suggests, in general, that efforts to identify sources of dioxin by examination of soil profiles must be considered suspect, since the dioxins apparently undergo transformations either in the air before reaching the soil or within the soil such that the profile may no longer be similar to the profile emitted into the air from nearby sources.

> *All congeners and homologue groups decreased in concentration when going from on-site to off-site soils, except for OCDD. The concentration of OCDD, in fact, increased substantially from on-site to off-site, only then to decrease with distance from the incinerator. No explanation for this trend could be found.*

A final observation of note for the soil monitoring is that all congeners and homologue groups are reduced in magnitude when going from on-site to off-site except OCDD. The concentration of this congener actually increases from 1,776 to 3,245 ppt. All other congener and homologue group concentrations drop by a factor of about 3 to greater than 10. This is a curious result whose explanation is not immediately known. If emission depositions occurred immediately from stack release, then soil concentrations would logically be highest near the incinerator. It seems reasonable that the exit velocity of the stack gas emissions would carry the emissions at least beyond the fenceline of the incinerator before substantial deposition occurs. This would be consistent with the hypothesis offered earlier that the three high on-site soil samples were influenced by ash drift.

> *A background urban air concentration of 3.5 pg/m^3 total and 0.05 pg TEQ/m^3 is hypothesized, based on an examination of the data. Two air samples known to be downwind of the operating Columbus WTE were higher, at average concentrations of 14.1 pg/m^3 total and 0.26 pg TEQ/m^3.*

Like the delineation of groups of soil samples, the air concentration data will be grouped, as follows: 1) "impacted air" - these are the two 1994 air samples found to be downwind of the Columbus WTE having the incinerator signature profile, 2) "1994 urban air" - these are the other 8 samples from 1994, 4 on each of two sample dates, 3) "1995 urban air" - these are the 6 urban air samples taken in 1995 after the Columbus WTE was shut down, and 4) "background air" - these are

the three air samples taken from the rural background site 45 km away - two samples in 1994 and one in 1995. Congener, homologue, total, and TEQ average concentrations are given for these groupings in Table 2.

Table 2. Congener-specific, homologue, total, and TEQ concentrations for the four clusters of air samples (units are pg/m^3) .

Congener	Impacted air (n=2)	1994 Urban (n=8)	1995 Urban (n=6)	Rural (n = 3)
2378-TCDD	0.019	0.003	0.007	0.003
12378-PCDD	0.062	0.012	0.008	0.005
123478-HxCDD	0.081	0.017	0.011	0.008
123678-HxCDD	0.095	0.028	0.024	0.009
123789-HxCDD	0.086	0.029	0.020	0.013
1234678-HpCDD	0.633	0.248	0.205	0.227
OCDD	1.765	1.062	0.807	0.904
2378-TCDF	0.051	0.012	0.017	0.003
12378-PCDF	0.121	0.024	0.022	0.007
23478-PCDF	0.169	0.028	0.020	0.010
123478-HxCDF	0.131	0.038	0.063	0.014
123678-HxCDF	0.302	0.056	0.058	0.016
123789-HxCDF	0.189	0.033	0.027	0.009
234678-HxCDF	0.006	0.003	0.003	0.003
1234678-HpCDF	0.939	0.165	0.165	0.061
1234789-HpCDF	0.131	0.027	0.038	0.014
OCDF	0.411	0.124	0.159	0.067
TCDD	0.761	0.097	0.110	0.015
PCDD	0.939	0.158	0.082	0.027
HxCDD	1.193	0.331	0.252	0.188
HpCDD	1.290	0.533	0.416	0.494
TCDF	1.793	0.374	0.378	0.083
PCDF	2.373	0.420	0.294	0.122
HxCDF	2.044	0.363	0.361	0.134
HpCDF	1.542	0.287	0.325	0.144
PCDD/PCDF	**14.11**	**3.75**	**3.18**	**2.18**
TEQ	**0.26**	**0.050**	**0.050**	**0.022**

The two impacted samples, with an average total concentration of 14.10 pg/m^3, are clearly elevated with respect to the other air samples while the group of three "rural air" samples, with an average total concentration of 2.18 pg/m^3, are lower as a group than the urban samples. As described in an earlier finding, the two impacted samples also had a profile similar to the stack gas emission profile. Although the 1994 urban samples had slightly higher average concentrations than did the 1995 urban samples, there was considerable overlap between the two groups. It is suggested that both of these groups represent an "urban background" ambient air concentration for the city of Columbus. This appears reasonable for 1995, since the incinerator was no longer operating at that time. Supporting evidence for the 1994 urban samples being representative of an urban background comes from the wind rose data presented in OEPA (1994b) for the two sample dates in 1994. That data shows that the wind was blowing mainly from the Northeast to the Southwest quadrants (and also from the Southwest to the Northeast), and therefore, emissions from the Columbus WTE would mainly effect only one sampler and possibly only marginally impact the other samplers; see Figure 1 identifying only one air sampler in the Southwest quadrant and none in the Northeast quadrant. Based on this evidence, the 1994 and 1995 urban sample sets suggest that the urban background air concentration is about 3.5 pg/m^3 total (specifically, 3.8 pg/m^3 for 1994 and 3.2 pg/m^3 for 1995) and 0.05 pg TEQ/m^3 (same TEQ for both years).

An examination of congener profiles in the air in 1995, after the Columbus WTE was no longer operating, suggests that there may be other incinerator signature sources impacting air quality in the Columbus urban area.

Although on average the 1995 urban air samples can be distinguished from the two "impacted air" samples of 1994, there are a few 1995 samples which have profiles similar to those of the Columbus WTE stack emissions and the two "impacted air" samples. Specifically, two urban 95 samples had OCDD percentages within 3% of stack average from the 1994 stack test: the stack emission average OCDD percentage was 16% - two 1995 ambient air samples had OCDD percentages of 19%. These two 1995 samples also had the highest air concentrations in the set of six 1995 urban samples, 4.8 and 4.7 pg/m^3 total compared to an average of 2.4 pg/m^3 total for the other four samples. It should be noted that neither of the two urban 95 samples with low OCDD percentages were from the same air monitor that produced the impacted air samples of 1994.

When moving from urban background air to rural background air, two trends are evident: the concentrations decrease and the profile changes with an enhancement of OCDD. When moving from urban background to rural background soil, the concentrations decrease but the profile is substantially unchanged. Also, when comparing background air and soil

profiles, the principal tendency is for OCDD to become more enhanced in the soil matrix.

Figure 6 shows the congener profiles for urban background air and soil, and for the rural background air and soil. The four predominant congeners, as noted earlier, include 1234678-HpCDD, OCDD, 1234678-HpCDF, and OCDF. As described in the finding above on the "incinerator signature" profiles, essentially all congeners except these four in all matrices contribute less than 1% to the profile (with the exceptions that 123478-HxCDF and 123678-HxCDF contribute 1.5 and 1.7%, respectively, to the urban air background profile). When going from urban background to rural background air, the percentage of OCDD in the profile increases from 27 to 42%. 1234678-HpCDD increases from about 6 to 10%, while the two furan congeners of note decrease by about 1-2%. Urban and rural background soil profiles are substantially similar, with small increases in the predominance of each of the four noted congeners by 1-3%.

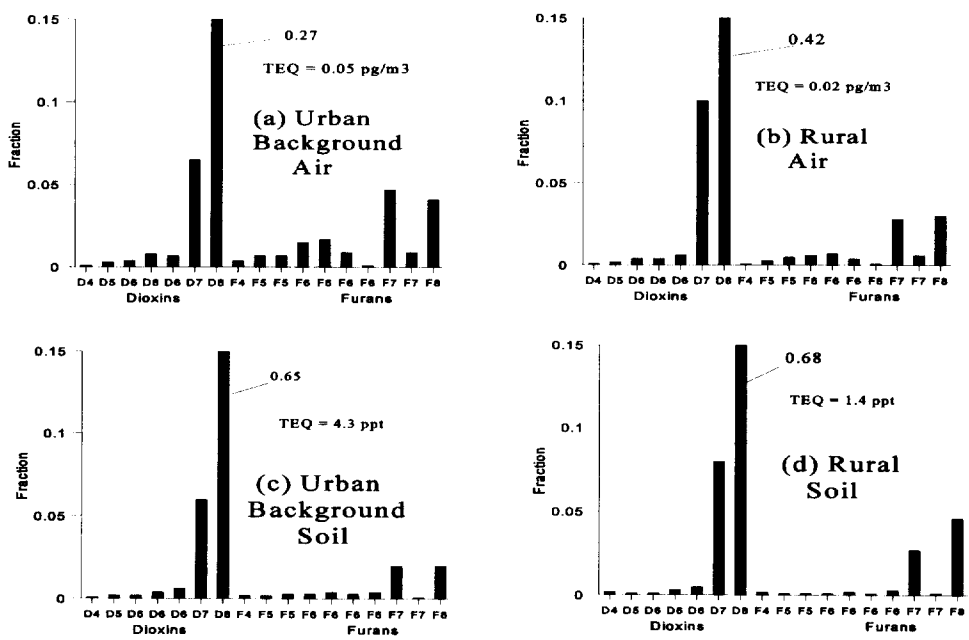

Figure 6. Background congener profiles for (a) urban background air averaging 1994 and 1995 profiles (not including the impacted air samples in 1994), (b) rural air, (c) urban background soil, and (d) rural soil (note: D4 = 2,3,7,8-TCDD; D5 = 1,2,3,7,8-PCDD; order of congeners on x-axis matches that of Tables 1 and 2).

In general, dioxins in air are thought to be the source of dioxins in soil. Exceptions include, for example, sludge-amended soils, ash landfills or soils near ash handling, highly contaminated soil

sites, soil amended by applications of pesticides containing dioxins, and so on. Ash drift may have contributed to the high dioxin concentrations for the "on-site" soil cluster, and other causes may have resulted in the high dioxin concentrations found in the city that were rejected from this study. Otherwise, depositions of dioxins from the air are expected to have been the cause of dioxins in the soil in the Columbus area. If that is the case, then Figure 6 suggests that the principal change in the profile from air to soil is that OCDD becomes enhanced when in the soil matrix. The most likely explanation is that OCDD, once on soil, becomes bound essentially where it deposits, with very little further environmental movement (volatilization, leaching) or degradation in comparison to all other congeners. Another possible explanation is that OCDD, thought to reside almost entirely on airborne particles, deposits proportionally more as compared to other congeners; conversely, other congeners which may have greater vapor fractions than OCDD have proportionally greater airborne transport than OCDD.

> *Analysis suggests that about 3 kg of total dioxins that were emitted from the incinerator remain in the soil in the impacted area near the incinerator. The stack test data suggests that between 150 and 550 kg of total dioxins were emitted from the Columbus WTE during its 11.5 years of operation. This suggests that less than 2% of emissions from the Columbus WTE can be found in soils near the incinerator.*

The curve-fit equation solving for soil concentration as a function of distance was, 13,000*exp (-1.75[distance]) + 514. This relationship was displayed in Figure 3. From this figure, it appears that background is about 3 km from the incinerator, with an average soil concentration at background at about 514 ppt. Therefore, this relationship, minus the +514 ppt term, can be used to estimate the average soil concentration within 3 km of the incinerator that is above background. By integration using polar coordinates of this equation between 0 and 3 km, it is shown that the average elevation in soil concentration within a circular area of 3 km is 913 ppt. Using the typical congener profiles for the urban and background soils (see Figure 5), this corresponds to approximately 7 ppt TEQ in elevation within 3 km. Figure 4 showing the isolines of equal soil concentration suggests that most of the impact is in the Northeast direction, but for the sake of conservatism, it will be assumed that this average pertains to a circular area (which will have an area defined as πr^2, with r = 3 km). Given a soil sampling depth of 7.5 cm and an assumed soil bulk density of 1.5 g/cm^3, this 913 ppt with the 3 km radius translates to a total of 2.9 kg as follows:

$$(913\ pg/g)\ (\pi\ 9\ km^2)\ (10^{10}\ \frac{cm^2}{km^2})\ (7.5\ cm)\ (1.5\ \frac{g}{cm^3})\ (10^{-15}\ \frac{kg}{pg})$$

Now, estimates can be made of the total emissions of dioxins and furans from the Columbus

WTE over its 11.5 years of operation using the 1992 and 1994 stack emission tests, coupled with the OEPA's (1994) assumptions regarding actual operation (in contrast to operating capacity). The estimates of total dioxin/furan emissions based on the 1992 stack test was 564 kg while the 1994 stack test was lower at 147 kg. Recall that the 1994 stack test is less likely to be representative of long term emissions since the test was conducted after process modifications reduced the emissions of dioxins. Still, given that an estimate of dioxins remaining in the soil is 2.9 kg, this suggests that between 0.5 and 2.0% of emissions deposited onto soils near the incinerator to result in elevations in concentrations.

The same analysis was done with individual congeners with similar results, with the exception of OCDD. Here, the percent of OCDD emitted which deposited in the impacted area was higher. Given that OCDD is about 65% of the total concentrations (see discussions above and Figure 5), than the mass of OCDD in the soil layer is about 1.9 kg. The emissions of OCDD were 18 and 29 kg from the 1992 and 1994 emission tests, respectively. Therefore, the percent of OCDD emitted which remained in the soil was between 6.6 and 10.6%.

The fate of the remaining emissions is, of course, not known. Among the possibilities include long range transport, environmental degradation, and leaching below the 7.5 cm depth of sampling.

Despite the magnitude of emissions from the Columbus WTE, they do not appear to have resulted in soil and ambient air concentrations in the urban area of Columbus which exceed urban soil and air concentrations of dioxins around the world.

The urban background TEQ soil concentration of about 4 ppt is somewhat low for typical urban settings. Other estimates of background urban TEQ in soil include a study in England showing a TEQ of 26 ppt (Ball, 1994) and a German study showing a range of concentrations in urban areas of between 10 and 30 ppt (BLAG, 1992, as reported in Fiedler, et al., 1995). However, only the cluster of 5 "off-site" samples within 1 km of the facility, averaging 49 ppt TEQ appear elevated above these urban soil concentrations. The average TEQ of 10 ppt found here in the "urban" cluster within 3 km of the WTE site does not appear to be out of the ordinary for urban settings, despite the high emissions from the Columbus WTE for over 11.5 years.

The rural samples have the lowest average soil concentration, 1 ppt TEQ, and these are typical of background soil concentrations. The data in rural settings in Germany showed a range of 1 to 5 ppt (BLAG(1994) as reported in Fiedler, et al. (1995)). In England, soil concentrations in a rural setting averaged 3.3 ppt TEQ (HMIP, 1995). Reed, et al.(1990) reports on a background soil concentration of 5.2 ppt TEQ in rural Minnesota. Fiedler, et al.(1995) reports on 36 soil samples taken in 8 counties in Southern Mississippi, in predominantly rural areas. The TEQ concentration

ranged from 0.08 to 22.9 ppt, with 20 samples being less than 1 ppt and a mean concentration of 3 ppt TEQ.

The urban air background concentration of 0.05 pg TEQ/m^3 also may be a little low for typical urban air concentrations. In a compilation of primarily urban United States air data, the United States Environmental Protection Agency calculated an average TEQ air concentration from 84 sample points of 0.09 pg TEQ/m^3 (EPA, 1994). The Columbus WTE did increase the urban air concentration in the city of Columbus, but it is unknown by how much. The two impacted air concentrations of 0.17 and 0.35 pg TEQ/m^3 were higher than the TEQ urban background of 0.05 pg TEQ/m^3 by about 3 to 7 times. Because of the small number of samples (sampling times and samplers), an accurate average urban air concentration during the years while the Columbus WTE was operating cannot be derived. Still, despite the magnitude of releases of dioxins from the Columbus WTE, it could be speculated that its overall impact to air quality appeared limited.

Summary and Discussion

This paper has examined the concentrations and profiles of dioxins in four matrices: stack emissions, ash, ambient air, and soil, all associated with the Columbus WTE. Short term impacts were noted in two ambient air samples with high concentrations and the retention of the incinerator signature. These two air samples were in the downwind direction less than two km away from the incinerator, and were taken while the incinerator was operating. No other samples of the same magnitude were taken in 1995, after the incinerator had shut down. Long term impacts were seen as elevated soil concentrations within about two km from the incinerator.

These elevated soil concentrations near the incinerator do indicate an important trend regarding the transformation of the incinerator signature. This signature is characterized by several congeners contributing a significant percentage to the overall profile (i.e., greater than 1%). However, the soil profile for this cluster of elevated soil samples have all but four congeners significantly reduced in relation to the four. These four congeners are: 1234678-HpCDD, OCDD, 1234678-HpCDF, and OCDF. What is additionally interesting is that the profile of this elevated cluster is found throughout the urban area sampled in Columbus all the way to the rural background site 45 km away, despite the fact that overall concentrations are declining through the city and at the rural site.

It would appear, therefore, that the stack emission profile eventually gets transformed to a soil profile. One theory offered to explain this is that OCDD, in particular, and the three other dominating congeners as well, have preferentially slower decay or dispersion rates compared to the other homologues and congeners. It has been found that vapor phase dioxins have a greater tendency to undergo degradation (photolysis, photooxidation) in sunlight as compared to particle

bound dioxins (EPA, 1994). This, coupled with the fact that the lower chlorinated dioxins tend to exist more in the vapor phase than the higher chlorinated dioxins, may partially explain why these four higher chlorinated congeners are accumulating in the soil. A related fate and transport consideration is that dry deposition, and possibly wet deposition, may be dominated by particle-phase rather than vapor-phase deposition. If that is the case, than the higher chlorinated dioxins, which, as just noted, tend to partition more in the particle phase compared to the lower chlorinated congeners, may deposit preferentially over the lower chlorinated dioxins.

This may explain the trend for soil concentration profiles outside the incinerator property, but it would not appear to explain the profile for three on-site soil samples which had the same signature profile as the stack emissions. The explanation offered earlier is that the onsite samples appear to have been impacted by drift of incinerator ash rather than deposition. However, this explanation would hold for all congeners except OCDD. In that case, the off-site concentration of OCDD is, in fact, substantially higher than the on-site OCDD concentration. This trend has no immediate explanation. As noted earlier, a possible explanation is that the exit velocity of the stack gas emissions would carry the emissions at least beyond the fenceline of the incinerator before substantial deposition occurs.

This evaluation of impacts near an incinerator emitting high amounts of dioxin is not the only evaluation of its kind. One other evaluation of impacts was conducted near a metals reclamation plant in Austria (Riss, et al., 1990; Riss, 1993). This plant was located in a rural Alpine river valley in Tyrol, Austria, in an area mostly comprised of agriculture. Although emissions data were unavailable, measured air concentrations near the incinerator of 1.2 -2.3 pg TEQ/m^3 (Riss, 1993) suggest very high emissions; recall that the high air concentrations noted near the Columbus incinerator were 0.17 and 0.35 pg TEQ/m^3. Soil concentrations averaged 420 pg TEQ/g at the site of the incinerator, 170 pg/g within 200 meters of plant, and 46 ppt about 2 km in the downwind direction (Riss, et al, 1990). This compares to an on-site soil concentration of 458 pg TEQ/g, and 49 pg TEQ/g within 2 km of the Columbus WTE. Sampling also occurred on a dairy farm at this Austrian site, which was located about 2 km meters from the site in the downwind direction, and members of that farming family consumed milk from their own cows. Elevated concentrations were found in cow's milk, vegetation, and blood levels from members of the family. In this case, the incinerator was still operating when the sampling occurred. These samples were taken in the late 1980s, before emission controls and other practices (i.e, removal of some plastics) were undertaken to reduce emissions from these plants. Riss (1993) reported on reductions in both cow's milk and fodder from this nearby farm in the early 1990s that he speculates to have resulted from reductions in incinerator emissions. Cows' milk dropped steadily from a high in 1987/88 samples averaging 49 pg TEQ/g fat to an average of 5 pg TEQ/g fat in 1992/93 sampling, while grass

similarly dropped from 33 pg TEQ/g dry weight to 4 pg TEQ/g dry weight between the two sample dates. This data was instrumental in decisions not to sample vegetation or biological tissue in 1995 after the Columbus WTE had shut down. This evidence demonstrates the relevance of the air-to-plant-to-animal (including the human animal) pathway when evaluating the near-field impacts of incinerator emissions of dioxins.

The final trend of note discussed above was that, despite evidence of high emissions over 11.5 yrs of operation, the overall air and soil concentrations in the Columbus urban area did not appear elevated in comparison to urban air and soil from other studies around the world. However, it would be inappropriate to conclude that single sources of dioxin release, no matter how high in magnitude, may not have appreciable impacts to the environment. Certainly, no information was gathered on the impact of emissions to vegetation or biota, and as indicated by the data from Austria, impacts could be noteworthy while emissions occur, but could then drop off rapidly once emissions are reduced or eliminated. An important observation was that less than 2% of the emissions could be found in the soil near the incinerator. This does not mean that 98% of the emissions did not influence the environment. Data suggests that dioxins are persistent in the environment, so it would appear that a large portion of the 98% of the remaining emissions continued to be dispersed beyond an area of immediate soil impact. It is possible, for example, that the background urban concentration of 4 ppt estimated in this analysis would instead be 2 ppt had the Columbus WTE never been in operation. Given the ubiquitous nature of dioxins in the environment, the only reasonable conclusion regarding the impact of the Columbus WTE is that it was widespread and far beyond the limits of the study area reported in this article.

ACKNOWLEDGMENT

The authors wish to thank Susan Perlin of the National Center of Environmental Assessment, USEPA, who used Arc/Info to draw Figures 1 and 4, and Lewis Summers of Lockheed Martin, who used the kriging option of Arc/Info to draw the contour lines shown in Figure 4.

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